Behavior of Electroplated Sn in Li/Sn cells

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Electrodeposition is an established inexpensive technology capable of fabricating novel materials at a fraction of the cost compared to physical vapor deposition methods (i.e.: sputter or vapor deposition). It is our goal to synthesize novel materials suitable for negative electrodes of Li-ion batteries via electrodeposition.

Tin was chosen as one component of the electrodeposit due to its large specific capacity. Our work began with a detailed study of electroplated tin. First, we learned to electroplate smooth tin layers that were tens of nanometers thick. When these were tested electrochemically in a Li battery their behaviour was unexpected. Large irreversible capacity plateaus were observed for a variety of deposition parameters and plating bath chemistries. Furthermore, the irreversible plateaus occur at odd stages in the charge-discharge cycle. Figure 1 shows a cell with large irreversible capacity in the first discharge, figure 2 shows a cell with an anomalous second discharge and figure 3 shows a cell with an anomalous third discharge. It is very odd to have such a distinct plateau present during the third discharge, with no presence in the first or second discharges. After an irreversible capacity plateau is observed the cell's capacity is dramatically decreased.

Besenhard et. al. [1] performed similar experiments on electroplated Sn. We duplicated the conditions of their deposits and also found large irreversible capacity in the early cycles as shown in figure 4. Our charge recovery data agrees with that published in [1], although there are no voltage profiles for the early cycles given in [1].

We have found some electrolyte dependence to the irreversible capacity; for example, it seems most evident when linear carbonates are used as co-solvents. Figure 5 shows results for a Li/Sn cell (electroplated Sn) using 1M $LiPF_6$/PC electrolyte. No anomalous plateaus are observed. We discuss five possible mechanisms to describe the irreversible behavior: water, Sn oxides, incorporation of plating bath additives, type of plating bath and catalytic decomposition of electrolyte. All but one, catalytic decomposition of electrolyte, can be eliminated. It is also demonstrated that cells having electrodes made from tin powder, carbon black and binder or electrodes of sputtered tin on copper foil, do not show any evidence of this irreversible capacity.

Reference
1. J. Yang, M. Winter and J.O. Besenhard, *Solid State Ionics* **90**, 281 (1996).

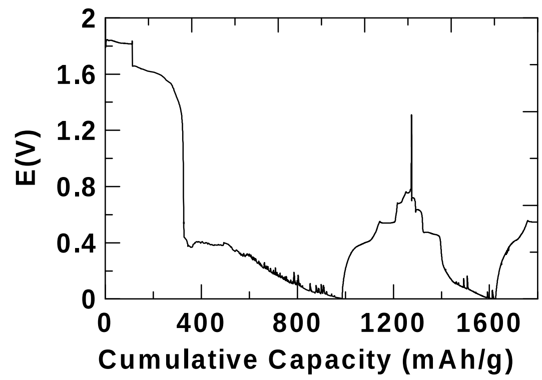

Figure 1. Voltage profile of a Li/Sn coin cell using electroplated Sn.

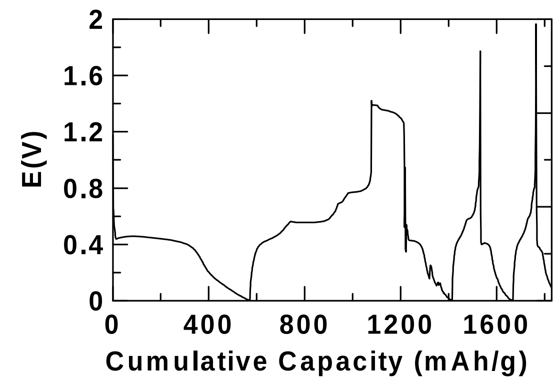

Figure 2. Voltage profile of a Li/Sn coin cell using electroplated Sn.

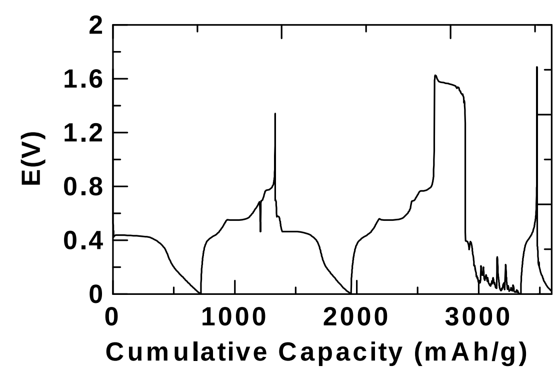

Figure 3. Voltage profile of a Li/Sn coin cell using electroplated Sn.

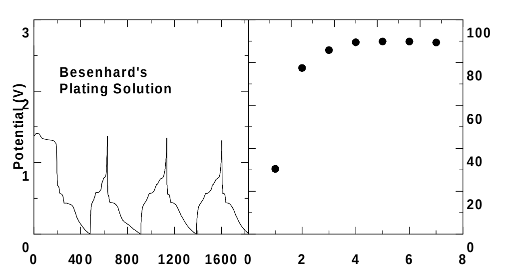

Figure 4. Voltage profile and charge recovery graph from a Li coin cell using the Sn plating chemistry from ref. 1. Our charge recovery data agrees with ref. 1.

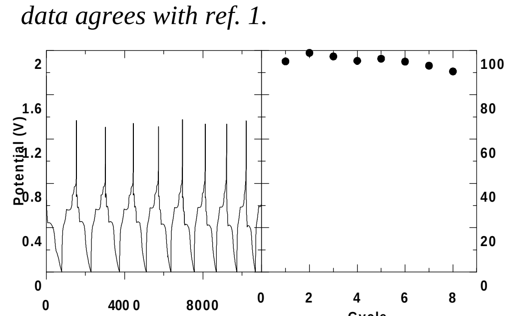

Figure 5. Voltage Profile of electroplated Sn in a Li cell using PC as the solvent.